

Application No. 09/843,814

Amendments to the Claims:

The following listing of claims will replace all prior versions, and listings, of claims in the application:

1. (Previously Presented) A catalytic composition comprising a solid support having deposited thereon a solid catalytically active material that is substantially insoluble in organic and aqueous liquid media, the solid catalytically active material consisting of at least one catalytically active anionic entity together with group 2 metal ions.
2. (Previously Presented) A catalytic composition as claimed in claim 1 wherein the solid catalytically active material is molecularly well defined.
3. (Previously Presented) A catalytic composition as claimed in claim 1 wherein the solid support includes external and pore surfaces, and pores that predominantly have a diameter greater than about 20 Å, and wherein the solid catalytically active material is deposited on the external and pore surfaces of the solid support.
4. (Previously Presented) A catalytic composition as claimed in claim 1 wherein the solid support includes pores that have pore diameters ranging from about 3 –3000 Å.
5. (Previously Presented) A catalytic composition as claimed in claim 1 wherein, the solid support is a chemically inactive solid material.
6. (Previously Presented) A catalytic composition as claimed in claim 1 wherein the solid support is porous and is powder, granules, flakes or pellets of regular or irregular shapes, sheets, monolith, ropes or woven fabric of fibrous solids.
7. (Previously Presented) A catalytic composition as claimed in claim 1 wherein the solid support is porous and is a thermally stable solid, insoluble in reaction media selected from the group consisting of organic, aqueous, fluorous, non-aqueous ionic liquids and supercritical fluid phases.

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8. (Previously Presented) A catalytic composition as claimed in claim 1 wherein the catalytically active anionic entity is insoluble in reaction media selected from the group consisting of organic, aqueous, fluorous, non-aqueous ionic liquids and supercritical fluid phases.

9. (Previously Presented) A catalytic composition as claimed in claim 1 wherein the solid catalytically active material is a thermally stable solid material having a melting point greater than 100°C.

10. (Canceled)

11. (Previously Presented) A catalytic composition as claimed in claim 1 wherein the solid support having deposited thereon the solid catalytically active material remains as a stable composite solid when used as a catalyst for gaseous, liquid or gas-liquid phase reactions.

12. (Previously Presented) A catalytic composition as claimed in claim 11 wherein the liquid phase is selected from the group consisting of organic, aqueous, fluorous, non-aqueous ionic liquids and supercritical fluid phases and mixtures thereof containing reactants, products or promoters.

13. (Previously Presented) A catalytic composition as claimed in claim 1 wherein the catalytic composition remains as a physically stable composite solid when used as a catalyst for gaseous or liquid phase reactions over a temperature range of 78 to 300°C.

14. (Previously Presented) A catalytic composition as claimed in claim 1 wherein the catalytic composition remains as a physically stable composite solid when used as a catalyst for gaseous or liquid phase reactions over pressures ranging from 5 to 5000 psi.

15. (Previously Presented) A catalytic composition as claimed in claim 1 wherein the group 2 metal ion is a cation having +2 charge.

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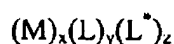
16. (Previously Presented) A catalytic composition as claimed in claim 1 wherein the group 2 metal is selected from the group consisting of calcium, strontium, barium and mixtures thereof.

17. (Previously Presented) A catalytic composition as claimed in claim 1 wherein the group 2 metal is selected individually or in combination with other group 2 metals.

18. (Currently Amended) A catalytic composition as claimed in claim 1 wherein the catalytically active anionic entity is an anion having, overall, a two or more negative charges charge of at least two.

19. (Previously Presented) A catalytic composition as claimed in claim 1 wherein the catalytically active anionic entity is individually selected from the group consisting of metal complexes, quaternary compounds, metaloxoanions, polyoxometallates and combinations thereof.

20. (Currently Amended) A catalytic composition as claimed in claim 19 wherein the catalytically active anionic entity is a metal complex having a general formula



wherein M is a metal atom or ion selected from the group consisting of group 3, 4, 5, 6, 7, 11 and 12 of the periodic table of elements, x ranges from 1 to 60, L is selected from the group consisting of aliphatic, aromatic and heterocyclic ligands containing at least one substituent selected from the group consisting of O, N, S, Se, Te, P, As, Sb, Bi, Si, olefin, and carbene that have attached thereto an oxy, alkyl, aryl, arylalkyl, alkylaryl, alkoxy, aryloxy, or cycloalkyl group bearing at least one or more negatively charged functional groups individually selected from the group consisting of $-\text{SO}_3^-$, $-\text{SO}_2^-$, $-\text{PO}_3^{2-}$, $-\text{COO}^-$, $-\text{O}^-$, AsO_3^{2-} and $-\text{S}^-$, y is at least 1, L^* is a ligand selected from the group consisting of an organic anion, an inorganic anion and coordinating compounds containing at least one substituent selected from the group consisting of O, N, S, Se, Te, P, As, Sb, Bi, Si, olefin, and carbene that have

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attached thereto an oxy, alkyl, aryl, arylalkyl, alkylaryl, alkoxy, aryloxy, or cycloalkyl group and z ranges from 0 to 7.

21. (Currently Amended) A catalytic composition as claimed in claim 19 wherein the catalytically active anionic entity is a quaternary compound having a general formula



wherein $l = 4$ for $Y^+ = N^+, P^+, As^+$; $l = 3$ for $Y^+ = S^+$ and R^* is selected independently from the group consisting of alkyl, aryl, arylalkyl, alkylaryl, alkoxy, aryloxy, and cycloalkyl bearing at least one or more negatively charged functional groups independently selected from the group consisting of $-SO_3^-$, $-SO_2^-$, $-PO_3^{2-}$, $-COO^-$, $-O^-$, AsO_3^{2-} and $-S^-$, and Z is an anion selected from the group consisting of an organic anion, an inorganic anion and a coordination complex anion.

22. (Previously Presented) A catalytic composition as claimed in claim 1 wherein the solid catalytically active material is deposited on the solid support along with at least one catalytically inert additive.

23. (Currently Amended) A catalytic composition as claimed in claim 22 wherein the catalytically inert additive is an anion having a two or more negative charges of at least two.

24. (Previously Presented) A catalytic composition as claimed in claim 22 wherein the catalytically inert additive is an anion, which is independently selected from the group consisting of organic anions, inorganic anions and combinations thereof.

25. (Currently Amended) A catalytic composition as claimed in claim 22 wherein the catalytically inert additive is selected from the group consisting of ligands that contain at least one substituent selected from the group consisting of O, N, S, Se, Te, P, As, Sb, Bi, Si, olefin, and carbene that have attached thereto an oxy, alkyl, aryl, arylalkyl, alkylaryl, alkoxy, aryloxy, or cycloalkyl group bearing at least one or more negatively charged functional groups

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independently selected from the group consisting of $-\text{SO}_3^-$, $-\text{SO}_2^-$, $-\text{PO}_3^{2-}$, $-\text{COO}^-$, $-\text{O}^-$, AsO_3^{2-} and $-\text{S}^-$.

26. (Previously Presented) A catalytic composition as claimed in claim 1 wherein an amount of the catalytically active anionic entity employed is 40 % weight or less of the catalytic composition.

27. (Previously Presented) A catalytic composition as claimed in claim 22 wherein an amount of the catalytically inert additive employed is in the proportion of 0 to 200 weight % of the catalytically active anionic entity.

28. (Canceled)

29. (Currently Amended) A catalytic composition as claimed in claim 1 further comprising a film of ~~high-boiling~~ liquid with a boiling point of less than 300°C deposited on the solid support.

30. (Previously Presented) A catalytic composition comprising a solid support having deposited thereon a solid catalytically active material that is substantially insoluble in organic and aqueous liquid media, the solid catalytically active material consisting of at least one catalytically active anionic entity and at least one catalytically inactive anionic additive that are together with group 2 metal ions.

31. (Previously Presented) A catalytic composition as claimed in claim 30 wherein the solid catalytically active material is molecularly well defined.

32. (Previously Presented) A catalytic composition as claimed in claim 30 wherein the solid support includes external and pore surfaces, and pores that predominantly have a diameter greater than about 20 Å, and wherein the solid catalytically active material is deposited on the external and pore surfaces of the solid support.

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33. (Previously Presented) A catalytic composition as claimed in claim 30 wherein the solid support includes pores that have pore diameters ranging from about 3 - 3000 Å.

34. (Previously Presented) A catalytic composition as claimed in claim 30 wherein the solid support is a chemically inactive solid material.

35. (Previously Presented) A catalytic composition as claimed in claim 30 wherein the solid support is porous and is powder, granules, flakes or pellets of regular or irregular shapes, sheets, monolith, ropes or woven fabric of fibrous solids.

36. (Previously Presented) A catalytic composition as claimed in claim 30 wherein the solid support is porous and is a thermally stable solid, insoluble in reaction media selected from the group consisting of organic, aqueous, fluoruous, non-aqueous ionic liquids and supercritical fluid phases.

37. (Previously Presented) A catalytic composition as claimed in claim 30 wherein the catalytically active anionic entity is insoluble in reaction media selected from the group consisting of organic, aqueous, fluoruous, non-aqueous ionic liquids and supercritical fluid phases.

38. (Previously Presented) A catalytic composition as claimed in claim 30 wherein the solid catalytically active material is a thermally stable solid material having a melting point greater than 100 °C.

39. (Canceled)

40. (Previously Presented) A catalytic composition as claimed in claim 30 wherein the solid support having deposited thereon the solid catalytically active material remains as a stable composite solid when used as a catalyst for gaseous, liquid or gas-liquid phase reactions.

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41. (Previously Presented) A catalytic composition as claimed in claim 40 wherein the liquid phase is selected from the group consisting of organic, aqueous, fluorous, non-aqueous ionic liquids and supercritical fluid phases and mixtures thereof containing reactants, products or promoters.
42. (Previously Presented) A catalytic composition as claimed in claim 30 wherein the catalytic composition remains as a physically stable composite solid when used as a catalyst in gaseous or liquid phase reactions over a temperature range of -78 to 300°C .
43. (Previously Presented) A catalytic composition as claimed in claim 30 wherein the catalytic composition remains as a physically stable composite solid when used as a catalyst in gaseous or liquid phase reactions over pressures ranging from 5 to 5000 psi.
44. (Previously Presented) A catalytic composition as claimed in claim 30 wherein the group 2 metal ion is a cation having +2 charge.
45. (Previously Presented) A catalytic composition as claimed in claim 30 wherein the group 2 metal is selected from the groups consisting of calcium, strontium, barium and mixtures thereof.
46. (Previously Presented) A catalytic composition as claimed in claim 30 wherein the group 2 metal is selected individually or in combination with other group 2 metals.
47. (Currently Amended) A catalytic composition as claimed in claim 30 wherein the catalytically active anionic entity is an anion having, overall, a two or more negative charges charge of at least two.
48. (Previously Presented) A catalytic composition as claimed in claim 30 wherein the catalytically active anionic entity is independently selected from the group consisting of metal complexes, quaternary compounds, metaloxoanions, polyoxometallates and combinations thereof.

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49. (Previously Presented) A catalytic composition as claimed in claim 30 wherein an amount of the catalytically active anionic entity employed is 40 % weight or less of the catalyst.

50. (Previously Presented) A catalytic composition as claimed in claim 30 wherein an amount of the catalytically inactive anionic additive employed is 0 to 200 weight % of the catalytically active anionic entity.

51. (Canceled)

52. (Currently Amended) A catalytic composition as claimed in claim 30 further comprising a film of ~~high-boiling~~ liquid with boiling point of less than 300°C deposited on the solid support.

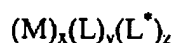
53. (Currently Amended) A process for preparing a catalytically active solid composition comprising a solid support, said process comprising reacting a solution consisting of a catalytically inactive additive and a catalytically active entity with a solution of group 2 metal cation and obtaining a precipitate,

wherein the catalytically inactive additive is independently selected from the group consisting of anions having a negative charge of at least two ~~or more negative charges~~, ligands containing at least one substituent selected from the group consisting of O, N, S, Se, Te, P, As, Sb, Bi, Si, and olefin that have attached thereto an oxy, alkyl, aryl, arylalkyl, alkylaryl, alkoxy, aryloxy, or cycloalkyl group bearing at least one or more negatively charged functional groups independently selected from the group consisting of $-\text{SO}_3^-$, $-\text{SO}_2^-$, $-\text{PO}_3^{2-}$, $-\text{COO}^-$, $-\text{O}^-$, AsO_3^{2-} and $-\text{S}^-$, and combinations thereof;

wherein the catalytically active entity is independently selected from the group consisting of metal complexes, quaternary compounds, metal oxo anions, polyoxometallates and combinations thereof,

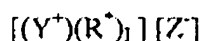
wherein the metal complexes have a general formula

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wherein M is a metal atom or ion selected from the group consisting of group 3, 4, 5, 6, 7, 11 and 12 of the periodic table of elements, x ranges from 1 to 60, L is selected from the group consisting of aliphatic, aromatic and heterocyclic ligands containing at least one substituent selected from the group consisting of O, N, S, Se, Te, P, As, Sb, Bi, Si, olefin, and carbene that have attached thereto an oxy, alkyl, aryl, arylalkyl, alkylaryl, alkoxy, aryloxy, or cycloalkyl group bearing at least one or more negatively charged functional groups independently selected from the group consisting of $-\text{SO}_3^-$, $-\text{SO}_2^-$, $-\text{PO}_3^{2-}$, $-\text{COO}^-$, $-\text{O}^-$, AsO_3^{2-} and $-\text{S}^-$, y is at least 1, L^* is a ligand selected from the group consisting of an organic anion, an inorganic anion and coordinating compounds containing at least one substituent selected from the group consisting of O, N, S, Se, Te, P, As, Sb, Bi, Si, olefin, and carbene that have attached thereto an oxy, alkyl, aryl, arylalkyl, alkylaryl, alkoxy, aryloxy, or cycloalkyl group, z ranges from 0 to 7 and

wherein the quaternary compound has a general formula



wherein, I = 4 for $Y^+ = \text{N}^+$, P^+ , As^+ ; I = 3 for $Y^+ = \text{S}^+$ and R^* is selected independently from the group consisting of alkyl, aryl, arylalkyl, alkylaryl, alkoxy, aryloxy, and cycloalkyl bearing at least one or more negatively charged functional groups independently selected from the group consisting of $-\text{SO}_3^-$, $-\text{SO}_2^-$, $-\text{PO}_3^{2-}$, $-\text{COO}^-$, $-\text{O}^-$, AsO_3^{2-} and $-\text{S}^-$ and Z is an anion selected from the group consisting of an organic anion, an inorganic anion and a coordination complex anion; and

wherein the group 2 metal cation is selected from the group consisting of Ca^{+2} , Sr^{+2} and Ba^{+2} ,

wherein the process further comprises depositing the precipitate upon the solid support.

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54. (Canceled)

55. (Previously Presented) A process as claimed in claim 53 wherein an amount of the catalytically active entity employed is 40 % weight or less of the catalytically active composition.

56. (Previously Presented) A process as claimed in claim 53 wherein an amount of the catalytically inactive additive employed is in the proportion of 0 to 200 weight % of the catalytically active entity.

57. (Canceled)

58. (Currently Amended) A process as claimed in claim 53 wherein a film of ~~high boiling~~ liquid having a boiling point of less than 300°C is deposited on the catalytically active composition.

59. (Currently Amended) A process for the preparation of a catalytic formulation as a solid composite comprised of a porous solid support having deposited thereon a catalytically active solid, said process comprising suspending the porous solid support in a liquid phase in which the porous solid support is insoluble to form a suspension, simultaneously adding to the suspension a solution of a catalytically inert additive and additive, ~~and~~ a catalytically active entity, and a solution of group 2 metal cation with sufficient agitation to maintain a suspension, and allowing to age for 1 to 48 hours,

wherein the porous solid support is a thermally stable solid in reaction media selected from the group consisting of organic, aqueous, fluorous, non-aqueous ionic liquids and supercritical fluid phases, has a mean pore diameter in the range of about 3-3000 Å and is in a form of powder, granules, flakes or pellets of regular or irregular shapes, sheets, monolith, ropes or woven fabric of fibrous solids,

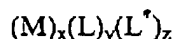
wherein the catalytically inert additive is independently selected from the group consisting of anions having a at least two or more negative charges ~~charge of at least two~~

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comprising organic anions, inorganic anions, or a compound containing at least one substituent selected from the group consisting of O, N, S, Se, Te, P, As, Sb, Bi, Si, olefin, and carbene that have attached thereto an oxy, alkyl, aryl, arylalkyl, alkylaryl, alkoxy, aryloxy, or cycloalkyl group bearing at least one or more negatively charged functional groups independently selected from the group consisting of $-\text{SO}_3^-$, $-\text{SO}_2^-$, $-\text{PO}_3^{2-}$, $-\text{COO}^-$, $-\text{O}^-$, AsO_3^{2-} and $-\text{S}^-$,

wherein the catalytically active entity is independently selected from the group consisting of metal complexes, quaternary compounds, metaloxoanions, polyoxometallates and combinations,

wherein the metal complexes have a general formula



wherein M is a metal atom or ion selected from the group consisting of group 3, 4, 5, 6, 7, 11 and 12 of the periodic table of elements, x ranges from 1 to 60, L is selected from the group consisting of aliphatic, aromatic and heterocyclic ligands containing at least one substituent selected from the group consisting of O, N, S, Se, Te, P, As, Sb, Bi, Si, olefin, and carbene that have attached thereto an oxy, alkyl, aryl, arylalkyl, alkylaryl, alkoxy, aryloxy, or cycloalkyl group bearing at least one or more negatively charged functional groups independently selected from the group consisting of $-\text{SO}_3^-$, $-\text{SO}_2^-$, $-\text{PO}_3^{2-}$, $-\text{COO}^-$, $-\text{O}^-$, AsO_3^{2-} and $-\text{S}^-$, y is at least 1, L^* is a ligand selected from the group consisting of an organic anion, an inorganic anion and coordinating compounds containing at least one substituent selected from the group consisting of O, N, S, Se, Te, P, As, Sb, Bi, Si, olefin, and carbene that have attached thereto an oxy, alkyl, aryl, arylalkyl, alkylaryl, alkoxy, aryloxy, or cycloalkyl group, z ranges from 0 to 7, and

wherein the quaternary compound has a general formula



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wherein, $I = 4$ for $Y^I = N^I, P^I, As^I$; $I = 3$ for $Y^I = S^I$ and R^I is selected independently from the group consisting of alkyl, aryl, arylalkyl, alkylaryl, alkoxy, aryloxy, and cycloalkyl bearing at least one or more negatively charged functional groups independently selected from the group consisting of $-SO_3^-$, $-SO_2^-$, $-PO_3^{2-}$, $-COO^-$, $-O^-$, AsO_3^{2-} and $-S^-$, and Z is an anion selected from the group consisting of an organic anion, an inorganic anion and a coordination complex anion and

wherein the group 2 metal cation is selected from the group consisting of Ca^{2+} , Sr^{2+} and Ba^{2+} .

60. (Canceled)

61. (Previously Presented) A process as claimed in claim 59 wherein the solution of the group 2 metal cation is selected from the group consisting of aqueous, water miscible organic and mixtures thereof.

62. (Previously Presented) A process as claimed in claim 59 wherein the solution of catalytically inert additive and catalytically active entity and the solution of group 2 metal cation are added simultaneously over a period of 10 to 1500 min.

63. (Previously Presented) A process as claimed in claim 59 wherein the solid composite is recovered by centrifugation, decantation or gravity settling, and is dried subsequently in a vacuum.

64. - 87. (Canceled).

88. (Currently Amended) A process for the preparation of a catalytic formulation as a solid composite comprised of a porous solid support having deposited thereon a catalytically active solid, said process comprising impregnating the porous solid support with a catalytically active entity and a catalytically inert additive followed by drying to obtain a dried porous solid support, adding the dried porous solid support having deposited thereon the catalytically active entity and the catalytically inert additive to a solution of a group 2 metal

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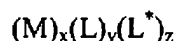
compound with simultaneous agitation, and aging the suspension for 1 to 48 hours with agitation,

wherein the porous solid support is a mechanically robust and thermally stable solid in reaction media selected from the group consisting of organic, aqueous, fluoruous, non-aqueous ionic liquids and supercritical fluid phases, has a mean pore diameter in the range of about 3-3000 Å and is in a form of powder, granules, flakes or pellets of regular or irregular shapes, sheets, monolith, ropes or woven fabric of fibrous solids and

wherein the catalytically inert additive is independently selected from the group consisting of anions having a at least two or more negative charges ~~charge of at least two~~ comprising organic anions, inorganic anions, or a compound containing at least one substituent selected from the group consisting of O, N, S, Se, Te, P, As, Sb, Bi, Si, olefin, and carbene that have attached thereto an oxy, alkyl, aryl, arylalkyl, alkylaryl, alkoxy, aryloxy, or cycloalkyl group bearing at least one or more negatively charged functional groups independently selected from the group consisting of $-\text{SO}_3^-$, $-\text{SO}_2^-$, $-\text{PO}_3^{2-}$, $-\text{COO}^-$, $-\text{O}^-$, AsO_3^{2-} and $-\text{S}^-$;

wherein the catalytically active entity is independently selected from the group consisting of metal complexes, quaternary compounds, metal oxo anions and polyoxometallates and combinations thereof,

wherein the metal complexes have a general formula



wherein M is a metal atom or ion selected from the group consisting of group 3, 4, 5, 6, 7, 11 and 12 of the periodic table of elements, x ranges from 1 to 60, L is an aliphatic, aromatic or heterocyclic ligand containing at least one substituent selected from the group consisting of O, N, S, Se, Te, P, As, Sb, Bi, Si, olefin, and carbene that have attached thereto an oxy, alkyl,

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aryl, arylalkyl, alkylaryl, alkoxy, aryloxy, or cycloalkyl group bearing at least one or more negatively charged functional groups independently selected from the group consisting of $-\text{SO}_3^-$, $-\text{SO}_2^-$, $-\text{PO}_3^{2-}$, $-\text{COO}^-$, $-\text{O}^-$, AsO_3^{2-} and $-\text{S}^-$, y is at least 1, L^* is a ligand selected from the group consisting of an organic anion, an inorganic anion and a coordinating compound containing at least one substituent selected from the group consisting of O, N, S, Se, Te, P, As, Sb, Bi, Si, olefin, and carbene that have attached thereto an oxy, alkyl, aryl, arylalkyl, alkylaryl, alkoxy, aryloxy, or cycloalkyl group, z ranges from 0 to 7 and

wherein the quaternary compound has a general formula



wherein $l = 4$ for $\text{Y}^+ = \text{N}^+$, P^+ , As^+ ; $l = 3$ for $\text{Y}^+ = \text{S}^+$ and R^* is selected independently from the group consisting of alkyl, aryl, arylalkyl, alkylaryl, alkoxy, aryloxy, and cycloalkyl bearing at least one or more negatively charged functional groups independently selected from the group consisting of $-\text{SO}_3^-$, $-\text{SO}_2^-$, $-\text{PO}_3^{2-}$, $-\text{COO}^-$, $-\text{O}^-$, AsO_3^{2-} and $-\text{S}^-$ and Z is an anion selected from the group consisting of an organic anion, an inorganic anion and a ~~or~~-coordination complex anion, and

wherein the group 2 metal cation is selected from the group consisting of Ca^{2+} , Sr^{2+} and Ba^{2+} .

89. (Canceled)

90. (Previously Presented) A process as claimed in claim 88 wherein the solution of the group 2 compound includes a solvent that is aqueous, water miscible organic or a mixture thereof.

91. (Previously Presented) A process as claimed in claim 88 wherein the porous solid support having deposited thereon the catalytically active entity and the catalytically inert additive is added to the solution of group 2 metal compound, with simultaneous agitation, over a period of 10 to 1500 min.

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92. (Previously Presented) A process as claimed in claim 88 wherein the solid composite is recovered by centrifugation, decantation, or gravity settling, and is dried subsequently in a vacuum.

93. (Currently Amended) A process for the preparation of a catalytic formulation as a solid composite comprised of a porous solid support having deposited thereon a catalytically active solid, said process comprising impregnating the porous solid support with a solution of a catalytically inactive additive and a catalytically active entity, followed by drying and suspending the porous solid support having deposited thereon the catalytically inactive additive and the catalytically active entity in a water immiscible solvent and adding a solution of a group 2 metal compound with agitation sufficient to maintain a suspension, and aging the suspension for 1 to 48 hours, and removal of low boiling or azeotropic fraction of solvent,

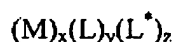
wherein the porous solid support is a mechanically robust and thermally stable solid in reaction media selected from the group consisting of organic, aqueous, fluoruous, non-aqueous ionic liquids and supercritical fluid phases, has a mean pore diameter in the range of about 70-3000 Å and is in a form of powder, granules, flakes or pellets of regular or irregular shapes, sheets, monolith, ropes or woven fabric of fibrous solids and

wherein the catalytically inactive additive is independently selected from the group consisting of anions having a at least two or more negative charges ~~charge of at least two~~ comprising organic anions, inorganic anions, or a compound containing at least one substituent selected from the group consisting of O, N, S, Se, Te, P, As, Sb, Bi, Si, olefin, and carbene that have attached thereto an oxy, alkyl, aryl, arylalkyl, alkylaryl, alkoxy, aryloxy, or cycloalkyl group bearing at least one or more negatively charged functional groups independently selected from the group consisting of $-\text{SO}_3^-$, $-\text{SO}_2^-$, $-\text{PO}_3^{2-}$, $-\text{COO}^-$, $-\text{O}^-$, AsO_3^{2-} and $-\text{S}^-$;

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wherein the catalytically active entity is independently selected from the group consisting of metal complexes, quaternary compounds, metal oxo anions and polyoxometallates and combinations thereof,

wherein the metal complexes have a general formula



wherein M is a metal atom or ion selected from the group consisting of group 3, 4, 5, 6, 7, 11 and 12 of the periodic table of elements, x ranges from 1 to 60, L is an aliphatic, aromatic or heterocyclic ligand containing at least one substituent selected from the group consisting of O, N, S, Se, Te, P, As, Sb, Bi, Si, olefin, and carbene that have attached thereto an oxy, alkyl, aryl, arylalkyl, alkylaryl, alkoxy, aryloxy, or cycloalkyl group bearing at least one or more negatively charged functional groups independently selected from the group consisting of $-\text{SO}_3^-$, $-\text{SO}_2^-$, $-\text{PO}_3^{2-}$, $-\text{COO}^-$, $-\text{O}^-$, AsO_3^{2-} and $-\text{S}^-$, y is at least 1, L^* is a ligand selected from the group consisting of an organic anion, an inorganic anion and a coordinating compound containing at least one substituent selected from the group consisting of O, N, S, Se, Te, P, As, Sb, Bi, Si, olefin, and carbene that have attached thereto an oxy, alkyl, aryl, arylalkyl, alkylaryl, alkoxy, aryloxy, or cycloalkyl group, z is from 0 to 7, and

wherein the quaternary compound has a general formula



wherein, I = 4 for $Y^+ = \text{N}^+$, P^+ , As^+ ; I = 3 for $Y^+ = \text{S}^+$ and R^* is selected independently from the group consisting of alkyl, aryl, arylalkyl, alkylaryl, alkoxy, aryloxy, and cycloalkyl bearing at least one or more negatively charged functional groups independently selected from the group consisting of $-\text{SO}_3^-$, $-\text{SO}_2^-$, $-\text{PO}_3^{2-}$, $-\text{COO}^-$, $-\text{O}^-$, AsO_3^{2-} and $-\text{S}^-$ and Z is anion selected from the group consisting of organic anion, inorganic anion and coordination complex anion, and

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wherein the group 2 metal compound comprises ions ~~is selected from the group consisting of compounds that include~~ Ca^{2+} , Sr^{2+} and Ba^{2+} .

94. (Canceled)

95. (Previously Presented) A process as claimed in claim 93 wherein a solvent employed to form a solution of the group 2 metal compound is aqueous, water miscible organic or a mixture thereof.

96. (Previously Presented) A process as claimed in claim 93 wherein the water immiscible organic solvent has a boiling point in a range of 40 to 200 °C.

97. (Previously Presented) A process according to claim 93 wherein the solid composite is recovered by centrifugation, decantation or gravity settling, and is dried subsequently in a vacuum.

98. (Currently Amended) A process for the preparation of a catalytic formulation as a solid composite comprising depositing a group 2 metal compound on a porous solid support followed by drying and suspending the porous solid support having deposited thereon the group 2 metal compound in a water immiscible solvent to which a solution of a catalytically active entity and a catalytically inactive additive is added with agitation sufficient to maintain a suspension, and removal of a low boiling or azeotropic fraction of solvent, and aging the suspension for 1 to 48 hours,

wherein the porous solid support is a thermally stable solid in reaction media selected from the group consisting of organic, aqueous, fluoruous, non-aqueous ionic liquids and supercritical fluid phases, has a mean pore diameter in the range of about 3-3000 Å and is in a form of powder, granules, flakes or pellets of regular or irregular shapes, sheets, monolith, ropes or woven fabric of fibrous solids and

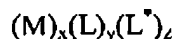
wherein the catalytically inactive additive is independently selected from the group consisting of anions having a ~~at least two or more negative charges~~ charge of at least two

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comprising organic anions, inorganic anions, ~~and or~~ a compound containing at least one substituent selected from the group consisting of O, N, S, Se, Te, P, As, Sb, Bi, Si, olefin, and carbene that have attached thereto an oxy, alkyl, aryl, arylalkyl, alkylaryl, alkoxy, aryloxy, or cycloalkyl group bearing at least one or more negatively charged functional groups independently selected from the group consisting of $-\text{SO}_3^-$, $-\text{SO}_2^-$, $-\text{PO}_3^{2-}$, $-\text{COO}^-$, $-\text{O}^-$, AsO_3^{2-} and $-\text{S}^-$;

wherein the catalytically active entity is independently selected from the group consisting of metal complexes, quaternary compounds, metal oxo anions and polyoxometallates and combinations thereof,

wherein the metal complexes have a general formula



wherein M is a metal atom or ion selected from the group consisting of group 3, 4, 5, 6, 7, 11 and 12 of the periodic table of elements, x ranges from 1 to 60, L is an aliphatic, aromatic or heterocyclic ligand containing at least one substituent selected from the group consisting of O, N, S, Se, Te, P, As, Sb, Bi, Si, olefin, and carbene that have attached thereto an oxy, alkyl, aryl, arylalkyl, alkylaryl, alkoxy, aryloxy, or cycloalkyl group bearing at least one or more negatively charged functional groups independently selected from the group consisting of $-\text{SO}_3^-$, $-\text{SO}_2^-$, $-\text{PO}_3^{2-}$, $-\text{COO}^-$, $-\text{O}^-$, AsO_3^{2-} and $-\text{S}^-$, y is at least 1, L^* is a ligand selected from the group consisting of an organic anion, an inorganic anion and a coordinating compound containing at least one substituent selected from the group consisting of O, N, S, Se, Te, P, As, Sb, Bi, Si, olefin, and carbene that have attached thereto an oxy, alkyl, aryl, arylalkyl, alkylaryl, alkoxy, aryloxy, or cycloalkyl group, z ranges from 0 to 7, and

wherein the quaternary compound has a general formula



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wherein $I = 4$ for $Y^I = N^+, P^+, As^+$; $I = 3$ for $Y^I = S^+$; and R^* is selected independently from the group consisting of alkyl, aryl, arylalkyl, alkylaryl, alkoxy, aryloxy, and cycloalkyl bearing at least one or more negatively charged functional groups independently selected from the group consisting of $-SO_3^-$, $-SO_2^-$, $-PO_3^{2-}$, $-COO^-$, $-O^-$, AsO_3^{2-} and $-S^-$, and Z is anion selected from the group consisting of an organic anion, an inorganic anion and a coordination complex anion and

wherein the group 2 metal compound comprises ions is selected from the group consisting of compounds that include Ca^{+2} , Sr^{+2} and Ba^{+2} .

99. (Canceled)

100. (Previously Presented) A process as claimed in claim 98 wherein a solvent employed to form a solution of the group 2 metal compound is aqueous, water miscible organic or a mixture thereof.

101. (Previously Presented) A process as claimed in claim 98 wherein the water immiscible organic solvent has a boiling point in a range of 40 to 200 °C.

102. (Previously Presented) A process as claimed in claim 98 wherein the solid composite is recovered by centrifugation, decantation, or gravity settling, and is dried subsequently in a vacuum.

103. (Currently Amended) A process for the preparation of a catalytic formulation as a solid composite comprising fluidizing a porous solid support in a flow of gases and spraying a solution of a catalytically active entity and a catalytically inert additive such that the catalytically active entity and the catalytically inert additive are deposited on the porous solid support, wherein the fluidizing is continued for 1 to 48 hours, and subsequently spraying a solution of a group 2 metal compound, and fluidizing is further continued for 1 to 48 hours, and recovering solids,

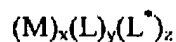
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wherein the porous solid support is a thermally stable solid in reaction media selected from the group consisting of organic, aqueous, fluorous, non-aqueous ionic liquids and supercritical fluid phases, has a mean pore diameter in the range of about 70-3000 Å and is in a form of powder, granules, flakes or pellets of regular or irregular shapes, sheets, monolith, ropes or woven fabric of fibrous solids and

wherein the catalytically inert additive is independently selected from the group consisting of anions having ~~a at least two or more negative charges~~ charge of at least two comprising organic anions, inorganic anions, ~~or and a compound containing at least one radical~~ selected from the group consisting of O, N, S, Se, Te, P, As, Sb, Bi, Si, olefin, and carbene that have attached thereto an oxy, alkyl, aryl, arylalkyl, alkylaryl, alkoxy, aryloxy, or cycloalkyl group bearing at least one or more negatively charged functional groups independently selected from the group consisting of $-\text{SO}_3^-$, $-\text{SO}_2^-$, $-\text{PO}_3^{2-}$, $-\text{COO}^-$, $-\text{O}^-$, AsO_3^{2-} and $-\text{S}^-$;

wherein the catalytically active entity is independently selected from the group consisting of metal complexes, quaternary compounds, metal oxo anions and polyoxometallates and combinations thereof,

wherein the metal complexes have a general formula



wherein M is a metal atom or ion selected from the group consisting of group 3, 4, 5, 6, 7, 11 and 12 of the periodic table of elements, x ranges from 1 to 60, L is an aliphatic, aromatic or heterocyclic ligand containing at least one substituent selected from the group consisting of O, N, S, Se, Te, P, As, Sb, Bi, Si, olefin, and carbene that have attached thereto an oxy, alkyl, aryl, arylalkyl, alkylaryl, alkoxy, aryloxy, or cycloalkyl group bearing at least one or more negatively charged functional groups independently selected from the group consisting of

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$-\text{SO}_3^-$, $-\text{SO}_2^-$, $-\text{PO}_3^{2-}$, $-\text{COO}^-$, $-\text{O}^-$, AsO_3^{2-} and $-\text{S}^-$, y is at least 1, L^* is a ligand selected from the group consisting of an organic anion, an inorganic anion and a coordinating compound containing at least one substituent selected from the group consisting of O, N, S, Se, Te, P, As, Sb, Bi, Si, olefin, and carbene that have attached thereto an oxy, alkyl, aryl, arylalkyl, alkylaryl, alkoxy, aryloxy, or cycloalkyl group, z ranges from 0 to 7, and

wherein the quaternary compound has a general formula



wherein $l = 4$ for $\text{Y}^+ = \text{N}^+$, P^+ , As^+ ; $l = 3$ for $\text{Y}^+ = \text{S}^+$ and R^* is selected independently from the group consisting of alkyl, aryl, arylalkyl, alkylaryl, alkoxy, aryloxy, and cycloalkyl bearing at least one or more negatively charged functional groups independently selected from the group consisting of $-\text{SO}_3^-$, $-\text{SO}_2^-$, $-\text{PO}_3^{2-}$, $-\text{COO}^-$, $-\text{O}^-$, AsO_3^{2-} and $-\text{S}^-$, and Z is an anion selected from the group consisting of an organic anion, an inorganic anion and a coordination complex anion, and

wherein the group 2 metal compound comprises ions ~~includes a cation~~ selected from the group consisting of Ca^{+2} , Sr^{+2} and Ba^{+2} .

104. (Canceled)

105. (Previously Presented) A process as claimed in claim 103 wherein the solvent employed to form a solution of the group 2 metal compound is aqueous, water miscible organic or a mixture thereof.

106. (Currently Amended) A process for the preparation of a catalytic formulation as a solid composite comprising tumbling a porous solid support in a rotating pan under a flow of gases, spraying a solution of a catalytically active entity and a catalytically inert additive such that the catalytically active entity and the catalytically inert additive are deposited on the porous solid support, the tumbling continuing for 1 to 48 hours, and

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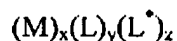
subsequently spraying a solution of a group 2 metal compound, and then tumbling is further continued for 1 to 48 hours, and solids are recovered,

wherein the porous solid support is a thermally stable solid in reaction media selected from the group consisting of organic, aqueous, fluoruous, non-aqueous ionic liquids and supercritical fluid phases, has a mean pore diameter in the range of about 70-3000Å and is in a form of powder, granules, flakes or pellets of regular or irregular shapes, sheets, monolith, ropes or woven fabric of fibrous solids and

wherein the catalytically inert additive is independently selected from the group consisting of anions having a at least two or more negative charges ~~charge of at least two~~ comprising organic anions, inorganic anions, or and a compound containing at least one substituent selected from the group consisting of O, N, S, Se, Te, P, As, Sb, Bi, Si, olefin, and carbene that have attached thereto an oxy, alkyl, aryl, arylalkyl, alkylaryl, alkoxy, aryloxy, or cycloalkyl group bearing at least one or more negatively charged functional groups independently selected from the group consisting of $-\text{SO}_3^-$, $-\text{SO}_2^-$, $-\text{PO}_3^{2-}$, $-\text{COO}^-$, $-\text{O}^-$, AsO_3^{2-} and $-\text{S}^-$;

wherein the catalytically active entity is independently selected from the group consisting of metal complexes, quaternary compounds, metal oxo anions and polyoxometallates and combinations thereof,

wherein the metal complexes have a general formula

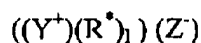


wherein M is a metal atom or ion selected from the group consisting of group 3, 4, 5, 6, 7, 11 and 12 of the periodic table of elements, x ranges from 1 to 60, L is an aliphatic, aromatic or heterocyclic ligand containing at least one substituent selected from the group consisting of O, N, S, Se, Te, P, As, Sb, Bi, Si, olefin, and carbene that have attached thereto an oxy, alkyl, aryl, arylalkyl, alkylaryl, alkoxy, aryloxy, or cycloalkyl group bearing at least one or more

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negatively charged functional ~~group groups~~ independently selected from the group consisting of $-\text{SO}_3^-$, $-\text{SO}_2^-$, $-\text{PO}_3^{2-}$, $-\text{COO}^-$, $-\text{O}^-$, AsO_3^{2-} and $-\text{S}^-$, y is at least 1, L^* is a ligand selected from the group consisting of an organic anion, an inorganic anion and a coordinating compound containing at least one substituent selected from the group consisting of O, N, S, Se, Te, P, As, Sb, Bi, Si, olefin, and carbene that have attached thereto an oxy, alkyl, aryl, arylalkyl, alkylaryl, alkoxy, aryloxy, or cycloalkyl group, z ranges from 0 to 7 and

wherein the quaternary compound has a general formula



wherein $I = 4$ for $\text{Y}^+ = \text{N}^+, \text{P}^+, \text{As}^+$; $I = 3$ for $\text{Y}^+ = \text{S}^+$ and R^* is selected independently from the group consisting of alkyl, aryl, arylalkyl, alkylaryl, alkoxy, aryloxy, and cycloalkyl bearing at least one or more negatively charged functional groups independently selected from the group consisting of $-\text{SO}_3^-$, $-\text{SO}_2^-$, $-\text{PO}_3^{2-}$, $-\text{COO}^-$, $-\text{O}^-$, AsO_3^{2-} and $-\text{S}^-$, and Z is anion selected from the group consisting of an organic anion, an inorganic anion and a coordination complex anion and

wherein the group 2 metal compound comprises ions ~~includes a cation~~ selected from the group consisting of Ca^{+2} , Sr^{+2} and Ba^{+2} .

107. (Canceled)

108. (Previously Presented) A process as claimed in claim 106 wherein a solvent employed to form the solutions is aqueous, water miscible organic or a mixture thereof.

109. (Previously Presented) A process as claimed in claim 106 wherein the solutions are sprayed simultaneously or sequentially.

110. (Canceled).